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Torrefaction and densification of different species of softwood residues

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HIGHLIGHTS

- Identify optimal torrefaction and densification conditions for torrefied pellet production.
- Quantify increased heating value and decreased moisture uptake for torrefied pellets.
- Quantify increased compression energy consumption and decreased density of torrefied pellets.
- Recommend high die temperature and sample preconditioning for torrefied pellet production.

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ABSTRACT

Torrefaction and densification of British Columbia (BC) softwoods, including pine, fir, spruce, SPF (a mixture of spruce, pine and fir) and pine bark, have been conducted for the production of high quality torrefied wood pellets. A bench-scale fixed bed tubular reactor was used for the torrefaction test at temperatures of 240–340 °C. Densification was conducted in a press machine in order to identify the suitable conditions for making strong torrefied pellets. Results showed that the mass loss of BC softwood mainly depended on the torrefaction temperature and time. The heating value of torrefied sawdust particles had a close relationship with the mass loss, increasing with increasing the severity of torrefaction. It was more difficult to compress torrefied samples into strong pellets than the raw material under the same conditions as used for making the control (regular and non-torrefied) pellets, and either a higher die temperature or adding moisture into torrefied particles could improve the compression process. The moisture content of torrefied pellets prepared in this study was lower than control pellets, and the density of torrefied pellets was slightly lower than control pellets. At the same time, more energy was consumed for compacting torrefied softwood particles into pellets. Increasing torrefaction severity increased the heating value and decreased the moisture uptake of torrefied pellets, but decreased the energy yield and the hardness of torrefied pellets. Considering the quality of torrefied pellets, the optimal torrefaction condition appeared to be around 30% mass loss, which gave a 20% increase in pellet higher heating value and a reasonable low water update rate. The suitable densification conditions for torrefied softwoods corresponded to a die temperature of 170–230 °C for unconditioned samples, or about 110 °C for samples preconditioned to ∼10% moisture content.

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1. Introduction

Torrefaction is a thermal treatment of wood at the temperature range of 200–300 °C without air or oxygen [1]. The major purpose of torrefaction in the biomass fuel industry is to increase the energy density on the mass basis and to improve the water resistivity of wood. A number of researches have been reported on biomass torrefaction in bench-scale units for different uses, such as co-firing in existing coal-fired power plants [2], gasification [3,4], syngas production [5], and barbeque fuel production [6]. Densification (also called pelletization) enhances the bulk density of wood residues from 40–200 kg/m^3 to 600–1400 kg/m^3 [7], which has been widely practiced in wood pellet plants. Reed and Bryant first considered the combination of torrefaction and pelletization to produce a new type of high energy density and water-resistant pellets [8]. Koukios observed apparent biomass energy densities exceeding 20 GJ/m^3 for torrefied straw, olive kernels and waste wood (softwood) pellets [9]. In Japan, the combined torrefaction and densification has been considered as an effective method to transport biomass from resources in remote areas to major urban centers [10].

The recent rapid growth of wood pellet industry has been mainly driven by the effort to reduce greenhouse gas emissions in Europe and the rising oil and natural gas prices. Bergman proposed and demonstrated a combined torrefaction and pelletization

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(TOP) process for the production of high energy density wood pellets [11]. Because of their high energy density and hygroscopicity properties, torrefied pellets are believed to be especially suitable for thermal power plants to replace coal and for export to Europe from North America over long transportation distances [12]. Currently, a number of torrefaction pilot plants have been designed, under construction, or publicly announced [13,14]. In Europe and Canada, two major joint programs have been recently initiated to demonstrate the production of torrefied pellets, with the first demonstration plant to be built in BC of Canada to produce 30,000 tonne/year torrefied pellets by 2015. So far, most researches have been focused on the development of torrefaction kinetics and reactors, with very few studies being conducted on densification of torrefied samples into pellets and combined torrefaction and densification. To build a commercial torrefied wood pellet plant, both torrefaction and densification technologies need to be developed, demonstrated and proved.

We previously developed a two-component first order one-step biomass torrefaction kinetic model based on the mass loss data from a thermogravimetric (TG) analyzer for the BC softwood to establish the relationship among the mass loss, torrefaction temperature and the residence time [15]. We also reported the particle size effect on torrefaction and densification [16], investigated the torrefaction of biomass residues in the presence of low oxygen concentrations [17], and studied the properties of torrefied wood pellets [18]. In this study, a bench-scale tubular reactor has been designed and built to study torrefaction and to produce enough torrefied wood samples for densification tests. A press machine, MTI 50K (Measurement Technology Inc.), was used to make and evaluate torrefied pellets from torrefied softwood samples. The main objective of this study is to identify the optimal conditions for combined torrefaction and densification for the production of high quality torrefied wood pellets.

2. Experimental

2.1. Samples

Pine and spruce chips from FPInnovations, fir and SPF (a mixture of spruce, pine and fir) chips from Fiberco, pine bark and SPF shavings from Wood Pellet Association of Canada, were used as biomass samples in this study. Woodchips and bark were prepared by drying with a THELCO laboratory PRECISION oven (Thermo Electron Corporation) at 105 °C for 24 h and crushing in a hammer mill (Glenmills Inc., USA; Model: 10HMBL) installed with different size screens. SPF shavings after milled with 4 mm opening screen were used for the study of temperature and residence time effects on torrefaction and densification. Spruce, fir, SPF, and pine bark sawdust samples were prepared using a hammer mill installed with a 0.79 mm opening screen, and then used for studying the effect of different BC softwood species on torrefaction and densification.

Table 1 shows the properties of BC softwood residues (ground materials). The volatile matter, fixed carbon, and ash content were measured by a TG analyzer (SHIMADZU, Japan; Model: TGA-50). The chemical composition was measured by FPInnovations using a high-performance liquid chromatography (Dionex Corporation, USA; Model: ICS-3000 Ion Chromatography System) following the ASTM E1758-01 procedure [19]. The elemental composition was determined by an elemental analyzer (Fisons, Germany; Model: EA 1108) in the Department of Chemistry at the University of British Columbia. The results in Table 1 show that the pine bark sample has a lower volatile mater content and a higher content in fixed carbon, ash, lignin and extractives than spruce, pine, fir and SPF samples. The elemental contents of spruce, pine, fir and SPF, on the other hand, were very close, but different from pine bark.

Table 1
Properties of BC softwood residues (bone dry material).

	Spruce	Pine	Fir	SPF	Pine bark	SPF shavings
Volatile matter (%)	80.5	82.8	82.9	84.4	75.8	85.1
Fixed carbon (%)	19.4	16.8	17.0	15.3	21.6	14.7
Ash (%)	0.1	0.4	0.1	0.3	2.6	0.2
Chemical composition						
Cellulose (%)	42.4	36.7	37.1	NA	26.1	NA
Hemicelluloses (%)	20.0	26.3	25.0	NA	17.9	NA
Lignin (%)	35.1	33.6	35.0	NA	41.0	NA
Extractives (%)	2.4	3.0	2.8	NA	12.4	NA
Elemental analysis (%)						
C	51.2	51.2	50.0	50.3	53.2	49.5
H	6.0	6.0	6.1	6.1	6.0	6.2
O (by difference)	42.6	42.2	43.6	43.2	37.7	43.9
N	0.1	0.1	0.2	0.1	0.5	0.2

Note: chemical composition of SPF is not measured because it depends on different fractions of spruce, pine and fir.

A THELCO laboratory PRECISION oven (Thermo Electron Corporation) was used to evaluate the moisture content for all biomass samples. A 25 ml glass cylinder was used to determine the bulk density [20]. A multipycnometer (Quantachrome Instruments, USA) was used to measure the particle density. The higher heating value was measured by a calorimeter (Parr 6100, USA). Particle size distributions were determined using a Ro-Tap sieve shaker (Tyler Industrial Products, USA). For the ground SPF shaving sample, the following sieves were used: 5, 7, 10, 14, 18, 25, 35, 45, 60, 80, 100, and 120 mesh (corresponding to 4.00, 2.80, 2.00, 1.41, 1.00, 0.707, 0.500, 0.354, 0.250, 0.177, 0.149, and 0.125 mm). For other samples, the following sieves were used: 18, 25, 35, 45, 60, 80, 100, 120, 170 and 230 mesh (corresponding to 1.00, 0.707, 0.500, 0.354, 0.250, 0.177, 0.149, 0.125, 0.088 and 0.063 mm). Sieving time was set at 5 min for each test run. The mass retained on each sieve was weighed to obtain the particle size distribution of the sample. In total, two replicates were measured for each sample. Fig. 1 shows the measured particle size distributions of different BC softwood species prepared with a grinder installed with a 0.79 mm size screen and a 4.00 mm size screen. From Fig. 1a, it is seen that the particle size distributions of prepared spruce, pine, fir and SPF samples were very close to each other, but different from the pine bark sample. Table 2 shows the properties of those samples. The particle density for all samples was around 1400 kg/m^3. The pine bark had a higher heating value than other samples. The mean sizes of spruce, pine, fir and SPF particles were very close to each other, but different from the pine bark sample. Those results indicated that the grinding behavior of pine bark could quite different from wood samples.

2.2. Torrefaction apparatus

A bench-scale fixed bed reactor was used for torrefaction tests. The unit includes a tubular reactor, an electrical gas pre-heater with a temperature controller and a power supply, a cooler, a condensate receiver, and a computer data acquisition system. The reactor has a inside diameter of 27 mm and a length of 575 mm, made from stainless steel. The reactor is located inside an electrically heated furnace with a reaction zone of ~100 mm in length. The biomass sample loaded into the reactor was heated up by both the preheated hot nitrogen gas flow and the electrical furnace. The hot nitrogen gas flows into the reactor from the top to the bottom. The temperature of the reactor zone was controlled by regulating the electric power via a temperature controller. The raw sample

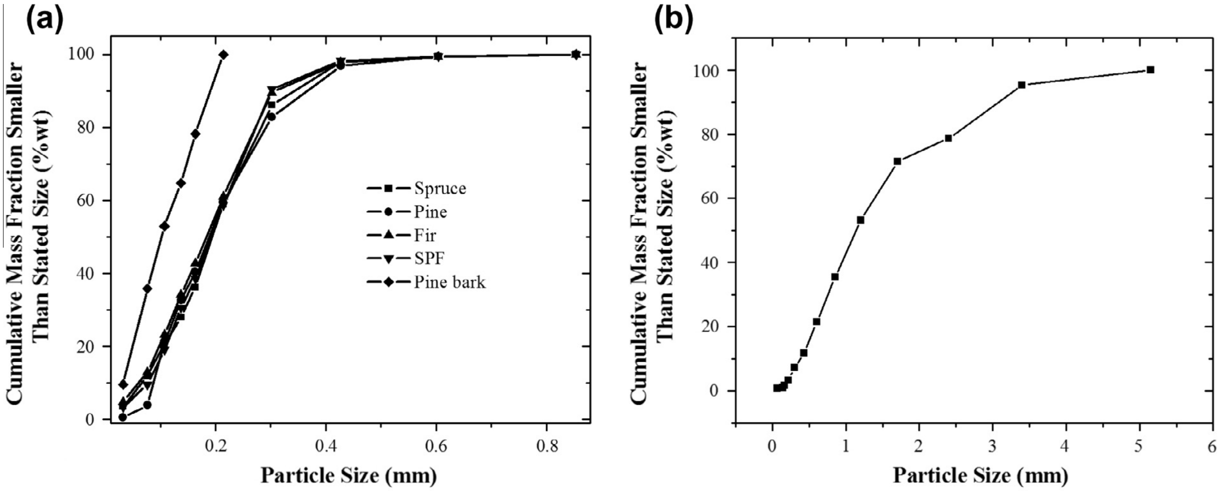

Fig. 1. Cumulative particle size distributions. (a) Different BC softwood samples prepared using a grinder with a 0.79 mm opening screen; and (b) SPF samples prepared using a 4.00 mm opening screen.

Table 2
Properties of BC softwood particle samples.

	0.79 mm Screen size					4.00 mm Screen size shavings
	Spruce	Pine	Fir	SPF	Bark	SPF
Moisture content (wt%)	9.5	7.4	3.2	3.5	7.2	9.9
Bulk density (kg/m^3)	200	230	230	180	310	160
Particle density (kg/m^3)	1410	1410	1440	1430	1380	1350
Higher heating value (MJ/kg)	18.3	18.6	18.4	18.6	19.5	18.1
Particle mean size (Sauter) (mm)	0.21	0.23	0.20	0.21	0.09	1.10

was added into the reactor from the top of the reactor, and the torrefied sample was discharged from the reactor after the reactor was cooled down following each run. All the condensable gases are collected in a water-cooled condensate receiver, and the non-condensable gases are collected by a gas sampling bag or discharged to a ventilation system (see Fig. 2). For each run, the unit was loaded with 20–40 g biomass samples.

During the torrefaction test, the nitrogen flow rate was set at 0.01416 N m^3/min. After the torrefaction, the flow rate increased to 0.02832 N m^3/min to cool down the torrefied sample. Four thermocouples are located in the unit to monitor the temperature. Thermocouple CH0 monitors the N_2 flow temperature. CH1 is for the biomass sample temperature. CH2 is for the gas heater surface temperature and CH3 is for the furnace temperature. The following procedures were followed in setting and controlling the reactor temperature: (1) preheating the nitrogen flow and the furnace to a temperature of 210 °C, which takes after 25 min; (2) setting up the furnace temperature to 30–40 °C above the target torrefaction

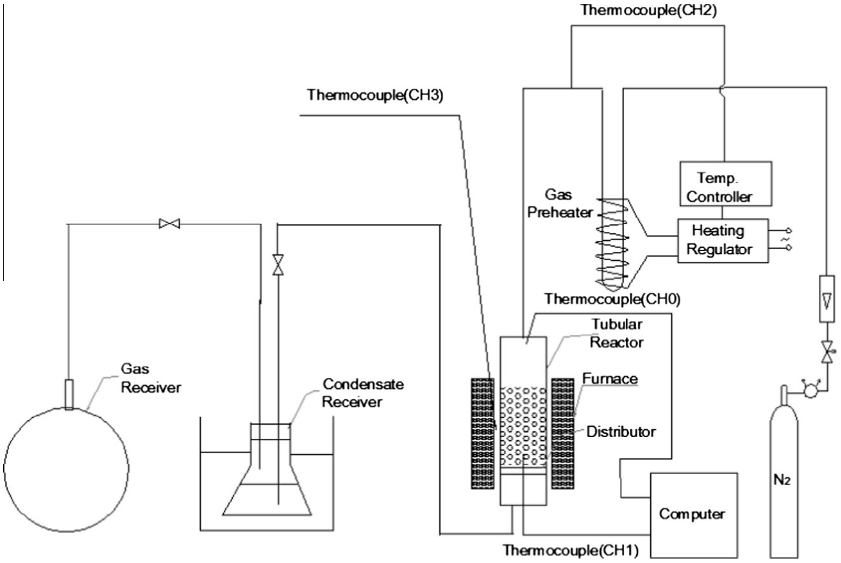

Fig. 2. Schematic of the fixed bed torrefaction test unit.

temperature, which enables the sample to be heated to the target torrefaction temperature over a few minutes; (3) controlling the furnace temperature to maintain the sample at the target torrefaction temperature for the preset residence time; (4) turning off the gas heater and furnace powers to cool the unit by keeping the nitrogen flow, until the sample temperature dropped to below 200 °C; (5) continuing to cool the unit for a few hours without nitrogen flow, until the sample temperature dropped to the room temperature before the sample was discharged. During the preheating period below 210 °C, it was checked that there was little decomposition of loaded samples in the reactor.

Two sets of tests were conducted. In the first set of tests, SPF shavings were torrefied at different torrefaction weight losses to investigate the effect of torrefaction severity on the properties of torrefied sawdust and pellets. In the second set, different species of softwood of similar particle size were torrefied with the same torrefaction weight loss, which was identified from the first set of tests, followed by densification, to examine the performance of different wood species. For each sample, at least two replicates were tested at each reaction condition.

2.3. Densification

A press machine (Measurement Technology Inc., USA; Model: MTI 50K) can be operated at various loading forces monitored by a load cell. A cylinder of 6.35 mm inside diameter and 70 mm in length with a piston 6.30 mm in diameter and 90 mm in length was installed to the machine for making a single pellet. The cylinder-piston unit was wrapped by a heating tape with a thermocouple and a temperature controller to preheat the cylinder to a certain temperature (also called die temperature). In this study, the cylinder was filled with approximately 0.5 g ground samples to make a single pellet of 6.5 mm in diameter and 12 mm in length. The sample was compressed by applying a pressure of 125 or 156 MPa to the die and then holding the die for a few minutes. The machine was then continued to press the sample until a maximum pressure of 156 or 187 MPa was reached. Because of the elastic relaxation, the pellet discharged out from the 6.35 mm die slightly expanded to 6.50 mm. Normally, for making control pellets, the die temperature was maintained at 70 °C with 125 MPa compression pressure and 1 min holding time [20].

Increasing the die temperature from 70 to 280 °C on compressing torrefied sawdust into pellets has been examined in this study, with the compression pressure ranging from 125 to 280 MPa. Preconditioning torrefied sawdust samples by adding 0%, 5%, 10% and 15% water and waiting for 24–72 h before compression was also explored. The effects of torrefaction conditions (e.g. temperature and weight loss) and wood species on densification were examined as well.

The single pellet density was calculated from measured mass and volume of individual pellets. Forces and piston displacements to form pellets were recorded to calculate the energy consumption. A Humidity Chamber (ESPEC CORP, Japan; Model: LHU-113) at 30 °C and 90% relative humidity was used for measuring the moisture uptake of torrefied and control pellets. The Meyer hardness (H_M) was applied to characterize the durability of alfalfa cubes with a flat surface [21]. The force was applied to indent the sample with a hemispherical probe, with the maximum force (F) to break a cube being recorded. The equation for calculating H_M for a hemispherical probe and a flat surface is given below:

$$H_M = \frac{F}{\pi(Dh - h^2)} \tag{1}$$

where D is the probe diameter and h is the indentation depth corresponding to the piston displacement before the cube breakage.

Lam et al. directly adapted Eq. (1) to the cylindrical pellets to obtain the hardness of pellets that was placed between two anvils under the MTI cross-head [22]. Wood pellets have a curved cylindrical surface. The cross-sectional area between the probe and the pellet is oval-shaped. Therefore, Eq. (1) needs to be modified as below when it is applied to the pellet:

$$H_M = \frac{F}{\pi\sqrt{Dh - h^2}\sqrt{\frac{D_p^2}{4} - \left[\frac{\frac{D_P^2}{2} + \frac{D \cdot D_P}{2} - D \cdot h - D_P \cdot h + h^2}{D + D_P - 2h}\right]^2}} \tag{2}$$

where D_p is the pellet diameter.

3. Results and discussion

3.1. Temperature gradient in the tubular reactor

Woody biomass has a very low thermal conductivity and heat capacity [23]. Since wood torrefaction is an endothermic reaction [3], these properties may create certain temperature gradients in the torrefaction reactor, which may affect the torrefaction performance.

To estimate the axial and radial temperature gradient in the tubular fixed bed reactor, torrefaction was assumed to be a simple one-step reaction with zero order and the reactor volume remains constant. The energy balance equation for torrefaction can then be simplified to:

$$\frac{\partial T}{\partial t} = \frac{k_s}{\rho_s C_p}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right)\right] - \frac{H_A k_0}{\rho_w C_P} e^{-\frac{E}{RT}} \tag{3}$$

where T is the temperature, K; t is the time, s; k_s is the thermal diffusivity of bulk pine sawdust; (estimated from $\frac{1}{k_s} = \frac{1-\varepsilon}{k_a} + \frac{\varepsilon}{k_w}$, k_w is the heat conductivity of pine, =0.109 W/(m K); k_a is the heat conductivity of air, =0.025 W/(m K), ε is the voidage of the packed bed of sawdust, =0.55) [22]; ρ_s is the bulk density of pine sawdust, =200 kg/m^3 [23]; C_p is the heat capacity of pine, =1640 J/(kg K) [23]; ρ_w is the particle density of pine sawdust, =440 kg/m^3 [24]; H_A is the heat of decomposition reaction, =792.15 J/g = 3.49 × 10^8 J/m^3 [25]; k_0 is the pre-exponential factor, =1.40 × 10^{10} 1/s [19]; E is the apparent activation energy, =150 kJ/mol [26]; R is the gas constant, 8.3144 J/(mol K).

The above differential equation was solved numerically by using the MATLAB software with proper initial and boundary conditions. The initial condition was set as: the temperature of sawdust was equal to 473 K at t = 0. The boundary conditions were set as: the wall surface temperature was constant at 573 K and $\frac{dT}{dr}\big|_{r=0} = 0$ at the center. The simulated results in Fig. 3 show that there exists a large radial temperature gradient in the reactor, with the temperature difference between the reactor wall and the center being more than 30 K after 500 s and remaining unchanged. It means that in the tubular reactor the temperature gradient can lead to the non-uniform torrefaction of sawdust along the radial direction.

The vertical temperature gradient was measured with a 10-point multi-thermocouple. The multi-thermocouple with a 48 mm interval was located at the center of the reactor tube to monitor the temperature profile. Fig. 4 shows the vertical temperature profiles in the reactor loaded with 0.2–0.3 mm diameter glass beads as a function of time. The furnace temperature was initially set to 340 °C. Fig. 4a shows that there exists a significant vertical temperature gradient, with a relatively uniform temperature zone located in the middle section of the reactor. To reduce the vertical and horizontal temperature gradient in the reactor, a hot N_2 flow was introduced. Fig. 4b shows the vertical temperature gradient profile in the reactor with 0.01416 N m^3/min hot N_2 flow.

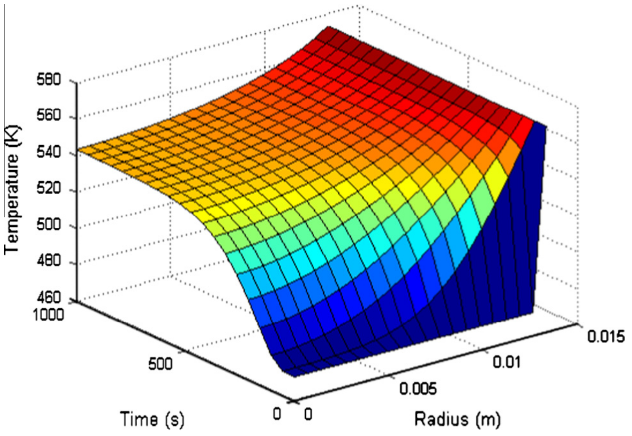

Fig. 3. Simulated radial temperature gradient profiles of pine sawdust in the packed bed unit.

As expected, the introduction of hot N_2 reduced the axial temperature gradient. Although not measured, the hot N_2 flow is also expected to reduce the radial temperature gradient significantly.

To achieve uniform torrefaction of biomass samples, both the radial and vertical temperature gradients should be minimized. In the current tubular reactor, a small reactor diameter should be used. In order to have enough samples prepared in each run for pelletization tests, a 27 mm diameter reactor tube was used in this study with a hot N_2 purge. Based on Fig. 4b, a relatively uniform temperature zone of ~100 mm long located in the middle section of the furnace was chosen as the torrefaction reaction zone, loaded with sawdust particles.

Fig. 5 shows the reactor temperature variation over time for a typical torrefaction run loaded with pine samples. The pre-heating from the room temperature to 200 °C took about 25 min with very little decomposition of biomass. It took about 10 min to further raise the reactor temperature from 200 to 250 °C, and 20 min to 300 °C. The sample cooling from the torrefaction temperature to below 200 °C took less than 10 min in order to terminate the torrefaction reaction. Fig. 5 also shows that at the beginning of torrefaction the furnace temperature needs to be controlled at 30–40 °C higher than the sample temperature to maintain the target torrefaction temperature of the sample because of the high endothermic torrefaction reaction rate. At the late stage with a low torrefaction rate, the temperature difference between the sample and the furnace became very small.

3.2. Torrefaction performance

Table 3 shows torrefaction results at temperatures of 240, 270, 300 and 340 °C with a residence time of 60 min for ground SPF samples, using the tubular reactor with continuous nitrogen purging. Torrefaction results for different BC softwood species, including spruce, fir, pine, SPF and pine bark, are given in Table 4. It is seen from both tables that as the temperature increased, the mass loss, the particle density, the higher heating value, and the carbon content all increased, but the bulk density, the energy yield, hydrogen and oxygen contents decreased. Those results are consistent with what were reported by Peng et al. and Wang et al. [16,17]. The higher particle density of torrefied sawdust than the control sawdust could be caused by the shrinkage of particles during torrefaction [27]. Because of the removal of water and oxygen-containing volatiles during torrefaction, the carbon content of the torrefied sawdust increased, giving rise to higher heating values of torrefied sawdust. At the same time, the removal of volatiles decreased the energy yield.

Fig. 6 plots the higher heating values against the mass loss for all torrefied samples, which revealed a strong correlation between them. The higher heating value increased steadily with increasing the severity of torrefaction, reaching 23 MJ/kg at a mass loss of 40%. For a 30% mass loss by torrefaction, the energy content increased by about 20%. From a least-square linear regression between the higher heating value and the mass loss data at mass losses less than 40%, the following relationship was obtained (excluding those data for the pine bark):

$$HHV_{\text{Torrefied Sample}} = (19.85 \pm 0.12) + (9.34 \pm 0.47) \times (\%\text{Mass Loss}) \quad (R^2 = 0.8862, N = 51) \qquad (4)$$

where 19.85 MJ/kg on the right hand side of the above equation corresponds to the average higher heating value of the dry raw material. For a first approximation, the higher heating value of torrefied biomass particles with a given mass loss could be estimated by Eq. (4).

Fig. 7 shows the van Krevelen diagram for untreated and torrefied BC softwood sawdust, including all tested data for dry raw materials and different torrefied materials. It is seen that both oxygen and hydrogen were removed selectively during torrefaction,

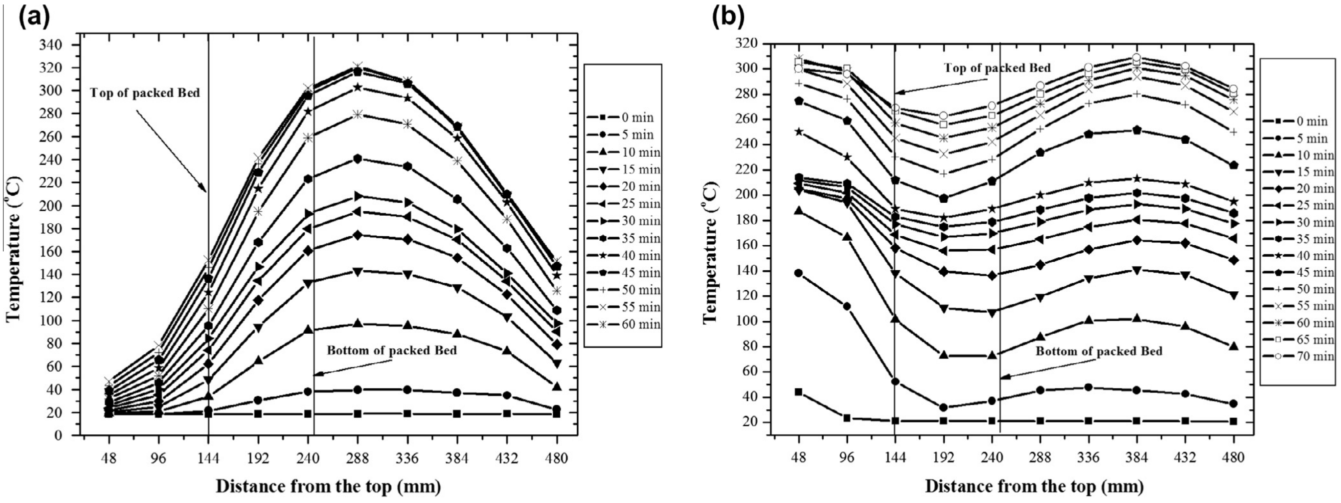

Fig. 4. Measured vertical temperature gradient profile in the 27 mm reactor filled with 0.2–0.3 mm glass beads. (a) Without N_2 flow; and (b) with 0.01416 N m^3/min hot N_2 flow.

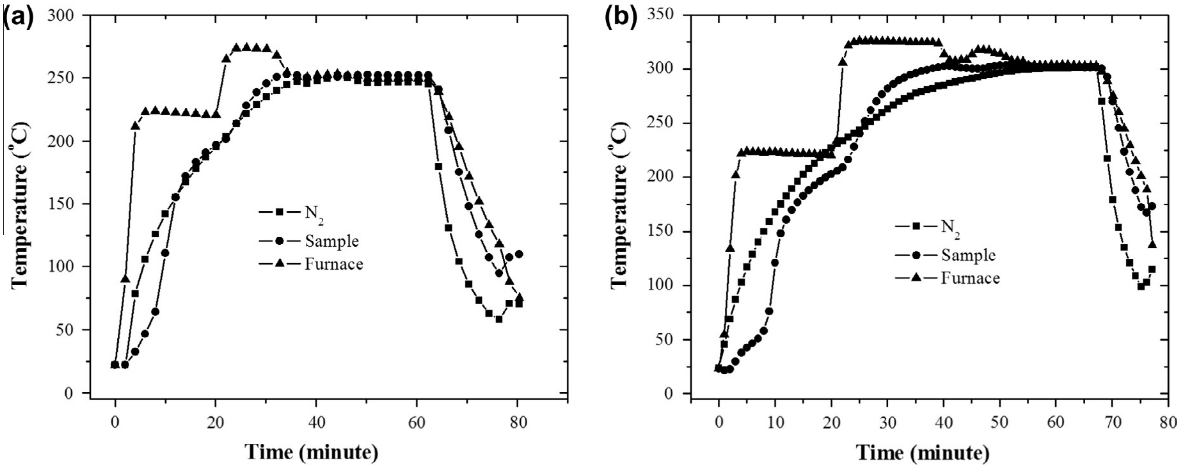

Fig. 5. Typical reactor temperatures as a function of time for the torrefaction reactor with pine samples. (a) 250 °C for 30 min; and (b) 300 °C for 30 min.

Table 3
Torrefaction results at different temperatures for SPF shavings.

Torrefaction temperature (°C)	240	270	300	340
Residence time, min	60	60	60	60
Bulk density (kg/m^3)	150	133	120	107
Particle density (kg/m^3)	1370	1360	1450	1450
Higher heating value (MJ/kg)	20.9	23.1	25.3	29.5
Mass loss (%)	13	34	51	61
Ash content (%)	0.2	0.3	0.4	0.5
Energy yield (%)	91	77	63	58
Elemental analysis (%)				
C	52.4	58.1	63.5	71.9
H	6.0	5.7	5.4	4.8
O (by difference)	41.2	35.7	30.5	22.6
N	0.2	0.2	0.2	0.2

Note: energy yield = (initial energy content of biomass – energy loss during torrefaction)/initial energy content of biomass.

Table 4
Torrefaction results of different BC softwood sawdust.

	Spruce	Pine	Fir	SPF	Pine bark
Particle mean size, mm	0.21	0.23	0.20	0.21	0.09
Torrefaction temperature (°C)	280	280	280	280	280
Residence time (min)	52	52	52	52	23
Bulk density (kg/m^3)					
Raw materials	200	230	230	180	310
Dry raw materials	190	190	220	160	250
Torrefied materials	160	180	190	140	230
Particle density (kg/m^3)					
Raw materials	1410	1410	1440	1430	1380
Dry raw materials	1450	1440	1460	1470	1410
Torrefied materials	1510	1530	1590	1600	1720
Higher heating value (MJ/kg)					
Raw materials	18.3	18.7	18.4	18.6	19.5
Dry raw materials	19.4	20.4	19.5	19.8	21.4
Torrefied materials	21.5	22.1	22.3	22.6	24.6
Mass loss (%)	24	29	27	31	30
Energy yield (%)	84	77	83	79	80
Ash content (%)	0.1	0.6	0.2	0.4	3.7
Elemental analysis (%)					
C	54.2	55.6	55.6	56.1	59.6
H	5.9	5.9	5.8	5.7	5.5
O (by difference)	39.81	37.8	38.3	37.7	30.5
N	0.1	0.1	0.1	0.1	0.7

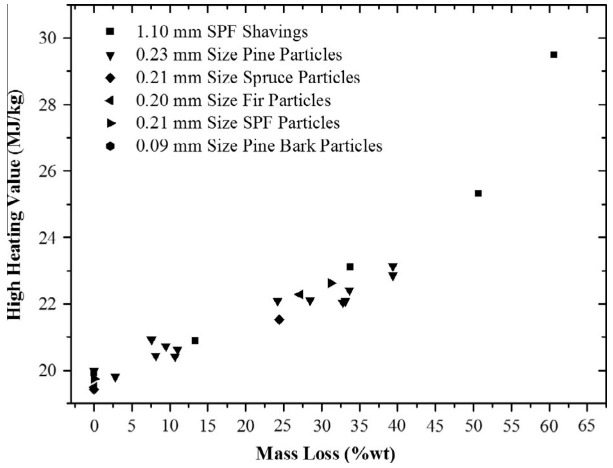

Fig. 6. Higher heating value of BC softwood samples as a function of mass loss.

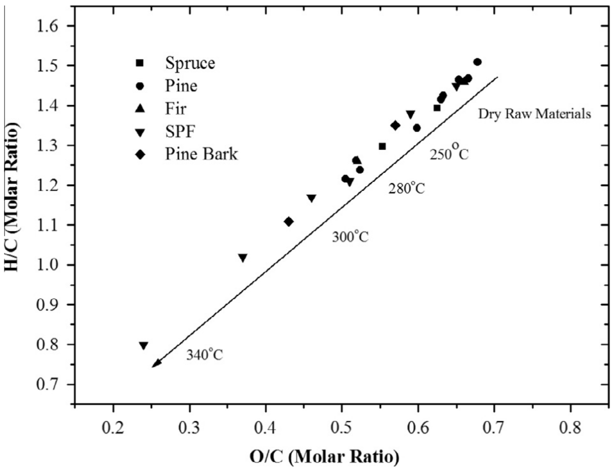

Fig. 7. van Krevelen diagram of untreated and torrefied BC softwood sawdust.

and they were closely correlated, as observed in pyrolysis. This could be explained by the selective removal of OH groups from the easy-to-decompose hemicelluloses during torrefaction. Similar results were reported by Madic et al. and Phanphanich and Mani [28,29]. A linear correlation between the H/C and O/C ratios for

Table 5
Summary of tested densification conditions.

Samples	Moisture content (%)	Die temperature (°C)	Pressure (MPa)	Numbers of pellets
Raw sawdust (0.23 mm pine)	7.4	70, 110, 170	31, 62, 125, 156	>50
Raw sawdust (0.21 mm spruce, 0.23 mm pine, 0.20 mm fir, 0.21 mm SPF, 0.09 mm pine bark)	3.2–9.9	70	125	>100
0.23 mm Torrefied pine sawdust and 1.10 mm SPF shavings	10 (added)	110	156	>200
Torrefied sawdust (0.23 mm pine at 280 °C for 52 min)	<0.5	170, 200, 230, 260, 280	125, 156, 187, 218, 249, 280	>50
Torrefied sawdust (0.21 mm spruce, 0.23 mm pine, 0.20 mm fir, 0.21 mm SPF, 0.09 mm pine bark)	<1	170, 230	156	>100
1.10 mm Torrefied SPF shavings	<1	170, 230	156	>20

Table 6
Properties of torrefied and control pellets made from BC softwood residues.

	Case 1	Case 2	Case 3	Case 4	Case 5
SPF Shavings					
Torrefaction temperature (°C)	Control	240	270	300	340
Residence time (min)		60	60	60	60
Die temperature, 170 °C					
Moisture content (%)	7.0	0.4	0.4	<0.1	0.2
Pellet density (kg/m^3)	1210	1100	1000	910	700
Energy consumption (MJ/t)	25.6	25.6	28.3	30.8	50.9
Meyer hardness (N/mm^2)	8.7	12.3	7.8	6.5	<1
Die temperature, 230 °C					
Pellet density (kg/m^3)	1210	1260	1200	1130	810
Energy consumption (MJ/t)	25.6	34.8	43.3	49.9	55.4
Meyer hardness (N/mm^2)	8.7	12.5	11.6	12.3	4.3
Different wood species	Spruce	Pine	Fir	SPF	Pine bark
Torrefaction Temperature (°C)	280	280	280	280	280
Residence time (min)	52	52	52	52	23
Control pellets					
Moisture content (%)	6.2	6.5	4.6	4.9	7.5
Pellet density (kg/m^3)	1170	1210	1140	1150	1290
Energy consumption (MJ/t)	29.1	27.5	31.4	31.2	18.7
Meyer hardness (N/mm$^{2)}$	10.2	10.4	4.5	5.4	18.8
Torrefied pellets					
Die temperature (170 °C)					
Moisture content (%)	0.2	0.5	0.3	0.3	0.8
Pellet density (kg/m^3)	1060	1080	1030	1010	980
Energy consumption (MJ/t)	31.2	30.6	33.3	35.2	28.1
Meyer hardness (N/mm^2)	7.7	7.8	7.2	7.1	4.7
Die temperature (230 °C)					
Moisture content (%)	0.2	0.6	<0.1	0.3	<0.1
Pellet density (kg/m^3)	1180	1220	1130	1150	1140
Energy consumption (MJ/t)	30.7	31.6	34.1	35.6	28.3
Meyer hardness (N/mm^2)	6.8	8.1	7.5	9.6	8.3

BC softwoods was obtained by least-square regression of experimental data:

$$H/C = (1.565 \pm 0.030) + (0.434 \pm 0.017) \times O/C \quad (R^2 = 0.9932, N = 21) \tag{5}$$

The relationship in Eq. (5) indicates that oxygen removal is faster than hydrogen removal from the biomass during torrefaction, likely because of the preferential extraction of oxygenated compounds to CO_2 and CO.

3.3. Densification characterization of torrefied pellets

Table 5 summarizes experimental conditions attempted to densify torrefied BC softwood sawdust into pellets. Torrefied sawdust samples were found to be more difficult to be compressed into strong pellets under the same operating conditions as used for making the control pellets from untreated sawdust, even at a die temperature of 150 °C [18,30,31]. The die temperature had to be increased to over 170 °C, or the samples had to be preconditioned to a moisture content up to 10% in order to be made into strong torrefied pellets [18].

Table 6 shows the properties of torrefied and control pellets made from different softwood residues. The moisture content of torrefied pellets was consistently lower than the control pellets because of the easy removal of unbonded free water from the torrefied sawdust during compression. The density of torrefied pellets was consistently lower than the control pellets when made under the same operating conditions, although the density of torrefied particles was higher than raw biomass particles. This is most likely due to the lowered elasticity of torrefied particles [18]. Li et al. also observed that the torrefied pellet density decreased with

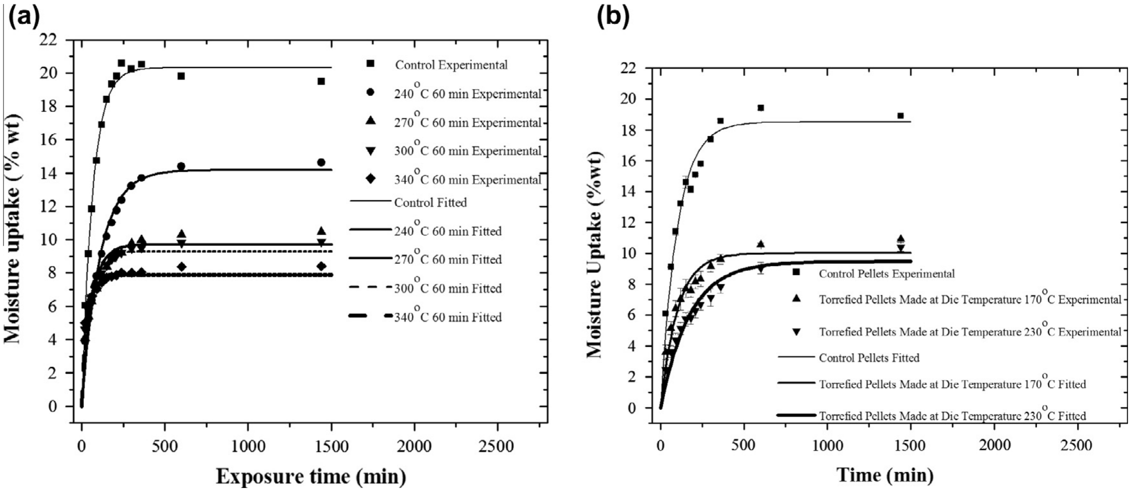

Fig. 8. Moisture uptake rate of control and torrefied pellets made from BC softwood residues. (a) 1.10 mm SPF shavings; and (b) 0.21 mm SPF particles at 280 °C for 52 min. (Pellets in a humidity chamber with 90% relative humidity air at 30 °C).

Table 7
Saturated moisture uptake and adsorption rate constant. (Pellets in a humidity chamber with 90% relative humidity air at 30 °C over 48 h).

	Mass loss (%)	Saturated moisture uptake, M_s (%)	Adsorption rate constant, k_m (min^{-1})
1.10 mm SPF shavings			
Control	0	20.3	0.015
240 °C 60 min	13	14.2	0.009
270 °C 60 min	34	9.7	0.017
300 °C 60 min	51	9.3	0.022
340 °C 60 min	61	7.9	0.039
Different wood species			
0.21 mm Spruce			
Control	0	19.1	0.007
280 °C 52 min Die 170 °C	24	10.2	0.010
280 °C 52 min Die 230 °C	24	9.7	0.006
0.23 mm Pine			
Control	0	18.6	0.006
280 °C 52 min Die 170 °C	29	10.7	0.008
280 °C 52 min Die 230 °C	29	9.6	0.006
0.20 mm Fir			
Control	0	18.6	0.011
280 °C 52 min Die 170 °C	27	10.7	0.011
280 °C 52 min Die 230 °C	27	9.9	0.007
0.21 mm SPF			
Control	0	18.5	0.010
280 °C 52 min Die 170 °C	31	10.0	0.010
280 °C 52 min Die 230 °C	31	9.5	0.006
0.09 mm Pine bark			
Control	0	18.2	0.004
280 °C 23 min Die 170 °C	30	8.6	0.015
280 °C 23 min Die 230 °C	30	8.4	0.011

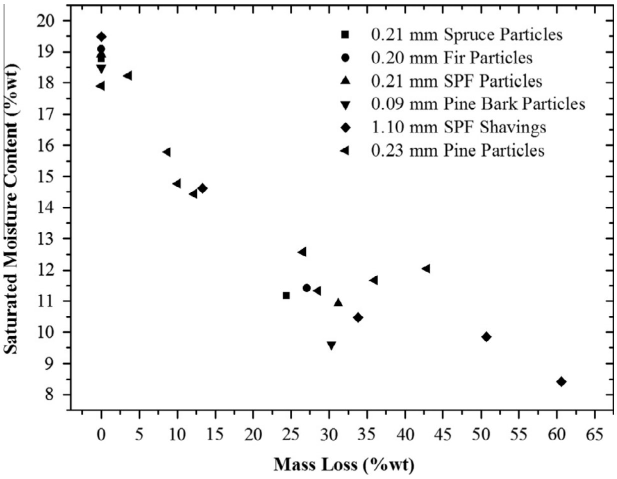

Fig. 9. Saturated moisture content as a function of torrefaction mass loss for all tested samples. (Pellets in a humidity chamber with 90% relative humidity air at 30 °C over 48 h).

increasing the mass loss and was lower than the control pellets [32]. Increasing the die temperature can improve the density of torrefied pellets significantly. Verhoeff et al. suggested that the die temperature had to be higher than 225 °C for making the high-density torrefied pellets [30]. At a die temperature of 230 °C, the density of torrefied wood pellets made from the 30% mass loss torrefied sawdust became similar to the density of control pellets. As the torrefaction temperature increased, the density of torrefied pellets decreased [18,32]. For sawdust torrefied at a temperature over 300 °C, both the hardness and the density of torrefied pellets became much lower. Both Li et al. and Peng et al. also reported that the hardness of torrefied pellets made at a die temperature of 170 °C was lower than control pellets [32,18]. A large compression force had to be applied for making pellets from torrefied SPF samples prepared at a torrefaction temperature of 340 °C. The energy consumption for making pellets also increased as the torrefaction temperature increased [18]. The energy consumption for making torrefied pellets from samples prepared at a torrefaction temperature of 300 °C was more than 50% higher than the control pellets. The Meyer hardness of torrefied pellets was comparable with the control pellets, although it was noticed that the torrefied wood pellets were more brittle than the control pellets. Based on the compression tests and pellet quality data (excluding pine bark), it appears to us that the optimal mass loss for making good torrefied softwood pellets should be around 30% prepared at a torrefaction temperature of 270–280 °C for about 55 min.

The moisture uptake of torrefied wood pellets was examined by measuring the water uptake rate in a humidity chamber with 90% relative humidity air at 30 °C over 48 h. Fig. 8 plots the measured moisture uptake rate of control and torrefied pellets made from BC softwood residues. It is seen from Fig. 8 that it took less than 10 h for most tested pellets to reach the saturated moisture

Table 8
Expansion of control and torrefied pellets made from BC softwoods after moisture uptake.

	Longitudinal expansion (%)	Diametric expansion (%)	Volumetric expansion (%)
1.10 mm SPF shavings			
Control	237	22	401
240 °C 60 min	67	14	118
270 °C 60 min	25	7	42
300 °C 60 min	14	9	35
340 °C 60 min	14	5	25
Different wood species			
0.21 mm Spruce			
Control	114	9	117
280 °C 52 min Die 170 °C	30	5	34
280 °C 52 min Die 230 °C	15	5	19
0.23 mm Pine			
Control	112	6	105
280 °C 52 min Die 170 °C	26	3	21
280 °C 52 min Die 230 °C	15	1	11
0.20 mm Fir			
Control	88	6	88
280 °C 52 min Die 170 °C	26	4	25
280 °C 52 min Die 230 °C	27	2	26
0.21 mm SPF			
Control	93	7	94
280 °C 52 min Die 170 °C	21	2	20
280 °C 52 min Die 230 °C	13	2	12
0.09 mm Pine bark			
Control	41	4	41
280 °C 23 min Die 170 °C	12	2	12
280 °C 23 min Die 230 °C	12	3	13

content. The saturated moisture content of control pellets was 19 ± 1 (wt%), and the saturated moisture content of all torrefied pellets was less than 10 (wt%), indicating substantial improvements in the hygroscopicity of torrefied wood pellets. The saturated moisture content of torrefied wood pellets also consistently decreased with increasing the degree of torrefaction in Fig. 8a because of the increased removal of hydroxyl (–OH) groups from the biomass [29,33]. Increasing the die temperature for pelletization, Fig. 8b, also slightly lowered the saturated moisture content of torrefied wood pellets.

The moisture uptake rate data can be fitted to the following first order kinetics equation as recommended by ASABE Standard S448.1 (2006):

$$\frac{M_m - M_s}{M_i - M_s} = \exp(-k_m t) \tag{6}$$

where M_m is the instantaneous moisture uptake, M_s is the saturated moisture uptake, M_i is the initial moisture uptake, k_m is the adsorption rate constant in min^{-1}, and t is the exposure time in min.

Table 7 shows the saturated moisture uptake (M_s) and the adsorption rate constant (k_m) estimated from fitting experimental data for control and torrefied pellets made at different torrefaction temperatures and densified at different die temperatures. M_s of torrefied pellets with ~30% mass loss ranged from 8% to 11%, about 50% lower than the control pellets (18–20%). Similar results were also reported by Peng et al. [18]. The adsorption rate constant k_m of torrefied pellets was in the same order as the control pellets, which might be explained by the increased specific surface area of torrefied pellets because of the lower pellet density and the creation of new pores inside the torrefied particles [18]. However, increasing the die temperature from 170 to 230 °C for pelletization significantly lowered the moisture adsorption rate constant, which might result from the increased pellet density and thermal modification of surface properties.

Fig. 9 shows the relationship between the saturated moisture content and the mass loss, or severity of torrefaction, for torrefied pellets. It is seen that the saturated moisture content of torrefied wood pellets decreased steadily with the increase in the mass loss or the degree of torrefaction. The decrease in saturated moisture content appears to be more significant at the weight loss of 0% to 15%, and stays steady afterward. A correlation between the saturated moisture content (M_s) and the mass loss for control and torrefied pellets was obtained by least-square regression of data:

$$\frac{M_s - (8.71 \pm 0.88)}{(18.87 \pm 0.31) - (8.71 \pm 0.88)} = \exp[(-0.0459 \pm 0.0094) \times \%\text{Mass Loss})] \quad (R^2 = 0.9555, N = 22) \tag{7}$$

The value of 18.87% most likely represents the average saturated moisture content of the raw material, while the value of 8.71% corresponds to the average saturated moisture content of charcoal [18].

Table 8 shows the diametric, longitudinal and volumetric expansions of control and torrefied pellets made from BC softwood residues after moisture uptake. It is seen from Table 8 that at the optimal mass loss of around 30%, the volumetric expansion of torrefied wood pellets made at a die temperature of 170 °C was around 30%. If the die temperature increased to 230 °C, the volumetric expansion of torrefied wood pellets with the same weight loss would decrease to around 10%. Comparing pellets made from two SPF samples of different particle sizes, the volumetric expansion of torrefied wood pellets made from small size particles was

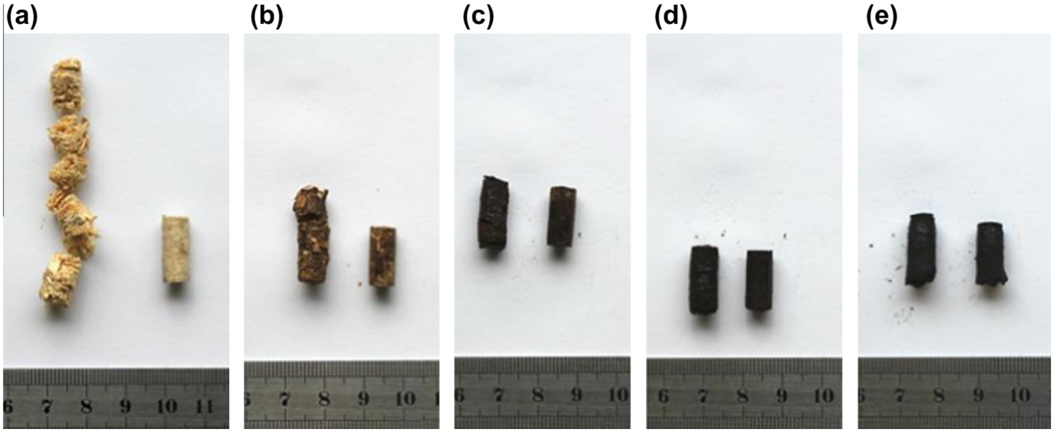

Fig. 10. Shape changes of control and torrefied pellets made from SPF shavings before and after moisture uptake. (a) Control pellets; (b) 13.3% mass loss (240 °C, 60 min); (c) 33.8% mass loss (270 °C, 60 min); (d) 50.7% mass loss (300 °C, 60 min); and (e) 60.6% mass loss (340 °C, 60 min).

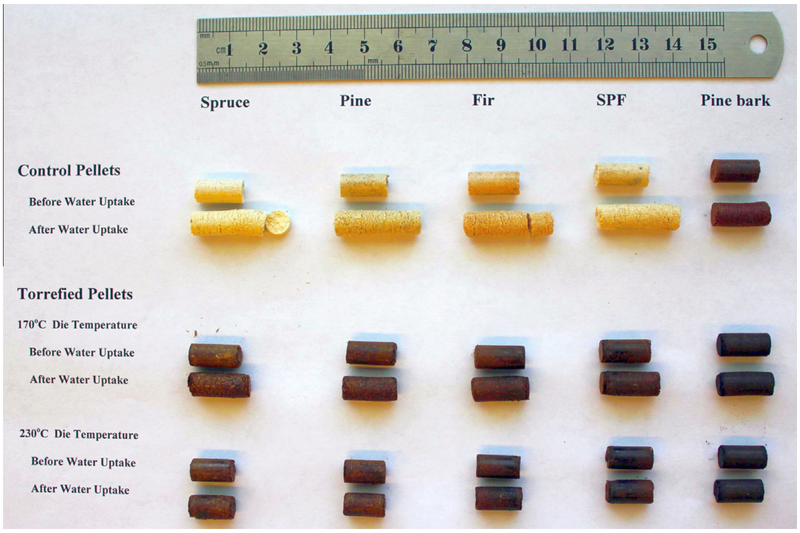

Fig. 11. Shape changes of control and torrefied pellets made from different softwood species after moisture uptake.

much lower than that made from large size particles. Deep torrefaction is seen to reduce the pellet expansion, and thus increase the quality of torrefied pellets. Increasing the die temperature also can improve the quality of torrefied pellets.

Fig. 10 shows the shape of the pellets before and after moisture uptake for control pellets and torrefied pellets, both made from SPF samples. On each picture, the pellet shown on the left was the one after moisture uptake, and the pellet on the right side was before the moisture uptake test. As seen in Fig. 10a, after the moisture uptake, the control pellet almost disintegrated. Pellets made from torrefied sawdust prepared at 240 °C for 60 min also showed a large expansion after the moisture uptake (see Fig. 10b). However, for those pellets made from torrefied samples with a mass loss of more than 30% (270 °C, 60 min; 300 °C, 60 min; 340 °C, 60 min), there was little change in the pellet shape and the pellets remained in good conditions after the moisture uptake. Fig. 11 shows the shape of the pellets before and after the moisture uptake for control pellets and torrefied pellets made from different BC softwood species. It is seen from Fig. 11 that at the high die temperature (260 °C) the expansion of torrefied pellets is very small after moisture uptake, and all torrefied pellets are seen to remain in their original shape after moisture uptake. Results shown in Figs. 10 and 11 seem to provide additional support to our previous recommendations that the optimal mass loss for the production of high quality torrefied pellets was around 30%.

4. Conclusions

The mass loss of British Columbia softwood species during torrefaction mainly depended on the reaction temperature and reaction time. The higher heating value of torrefied softwood had a strong correlation with the mass loss, with the higher heating value of torrefied sawdust at higher degree of torrefaction.

Torrefied softwood was more difficult to be compressed into strong pellets than control pellets under the same densification conditions. Increasing the die temperature and adding moisture into torrefied sawdust can improve the quality of torrefied pellets, but more energy was still needed for compacting torrefied sawdust into pellets. The saturated moisture content and density of torrefied pellets were lower than control pellets. Deep torrefaction increased the higher heating value and the hygroscopicity of torrefied pellets, but decreased the energy yield and the hardness of torrefied pellets. Torrefied pellets have a similar moisture adsorption rate as the control pellets, but their saturated moisture contents are at least 50% lower than the control pellets at a given moist environment.

Considering the quality of torrefied pellets, the optimal torrefaction conditions appeared to correspond to a mass loss of around 30%, which gave a 20% increase in pellet higher heating value. The suitable densification conditions for torrefied softwood based on the current study corresponded to a die temperature of 170–230 °C, and lower die temperatures could be used when the torrefied sawdust was preconditioned to a moisture content of around 10%.

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